

Conforming the measured lifetimes of the $5d\ ^2D_{3/2,5/2}$ states in Cs with theory

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We find very good agreement between our theoretically evaluated lifetimes of the $5d\ ^2D_{3/2}$ and $5d\ ^2D_{5/2}$ states of Cs with the experimental values reported in [Phys. Rev. A **57**, 4204 (1998)], which were earlier evinced to be disagreeing with an earlier rigorous theoretical study [Phys. Rev. A **69**, 040501(R) (2004)] and with another precise measurement [Opt. Lett. **21**, 74 (1996)]. In this work, we have carried out calculations of the radiative transition matrix elements using many variants of relativistic many-body methods, mainly in the coupled-cluster theory framework, and analyze propagation of the electron correlation effects to elucidate their roles for accurate evaluations of the matrix elements. We also demonstrate contributions explicitly from the Dirac-Coulomb interactions, frequency independent Breit interaction and lower order quantum electrodynamics (QED) effects. Uncertainties to these matrix elements due to different possible sources of errors are estimated. By combining our calculated radiative matrix elements with the experimental values of the transition wavelengths, we obtain the transition probabilities due to both the allowed and lower order forbidden channels. Adding these quantities together, the lifetimes of the above two states are determined precisely and plausible reasons for the reported inconsistencies between the earlier theoretical calculations and the experimental results have been pointed out.

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I. INTRODUCTION

Alkali atoms in general and Cs atom in particular are very interesting for many important experimental studies owing to their simple atomic energy level spacings and being suitable for optical magnetometry that are adequate to set up instrumentation for carrying out sophisticated measurements [1, 2]. These atoms couple extremely weakly to the environment allowing potentially very long coherence times. Few such categorical experiments are quantum computing [3, 4], observing parity nonconservation (PNC) effects to probe new particle physics [5, 6] and exotic property like nuclear anapole moment (NAM) [7, 8], measuring electric dipole moment (EDM) due to parity and time reversal symmetry violations [9, 10] etc.. In most of these studies, roles of accuracies in the theoretical calculations of various atomic properties are of also crucial [8, 10–15]. In fact, one of the reasons why experiments on the alkali atoms are so popular because many calculations of the ground and excited states properties are already performed that are as precise as the experimental results. This provides good test of validity of both the experimental and theoretical results and entrust confidence to infer many fundamental physics by combining these results [7, 8, 10–12]. However, it is still found conflicts between the theoretical and experimen-

tal results of these atoms in few cases; especially while studying properties of the excited states. One of such examples is the disagreements between the calculations and experimental results of the lifetimes of the $5d\ ^2D_{3/2}$ and $5d\ ^2D_{5/2}$ states of Cs [13]. Again, there are at least two precise measurements have been carried out to determine the lifetimes of the $5d\ ^2D_{3/2}$ and $5d\ ^2D_{5/2}$ states of Cs [16, 17] among which the lifetime of the $5d\ ^2D_{5/2}$ state has been reported as 1281(9) ns [16] and 1225(12) ns [17]; which are clearly out of the ranges of their reported error bars. The difficult part to perform precise calculations of the excited state properties lies in the strong roles played by the continuum in the evaluation of the atomic wave functions. Moreover determination of wave functions of the excited states with large orbital angular momentum (l), e.g. D states, demand large configuration state functions (CSFs) for which a method like configuration interaction (CI) method approximated only at the singles and doubles excitations (CISD method) may not be suitable enough to estimate their properties more accurately [18, 19]. On the otherhand, relativistic coupled-cluster (RCC) method even at the same levels of singles and doubles approximation (CCSD method) seems to be capable of estimating many atomic properties within the reasonable accuracies [20–22]. One of the unique features of the (R)CC method is, it can capture higher excited CSFs even approximating the method with the singles and doubles excitations owing to its ansatz of expressing the wave functions in the exponential form [22–25]. For example, the CCSD method still accounts contributions

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from most of the triples, quadruples etc. CSFs through the non-linear terms [24, 25]. Attaining precise results for the excited states with higher l values, inclusion of the contributions from these triples and quadruples CSFs are imperative. However, the CCSD method with only the linear terms (LCCSD method) in addition with other corrections is often being employed in the atomic wave function calculations of the alkali atoms due to requirement of large computational resources for accounting for the non-linear RCC terms (e.g. see Ref. [13] and references therein). Thus, comparative theoretical and experimental studies of the excited state atomic properties of the alkali atoms using a method would serve as a good test of its capability.

This work is intended to probe again the validity of the reported experimental values of the lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states of Cs which, as mentioned above, are inconsistently reported in different works [13]. Though experimental values from Refs. [16, 17] are discussed in Ref. [13], but few more measured values are also available in the literature with large error bars [26–28]. Similarly few more calculations of lifetimes of the above states are also known in the literature [29–33], however many of them are estimated using the non-relativistic theory and semi-empirical approaches. To appreciate consideration of the extra physical effects in the present work and to realize the reason for observing inconsistencies among the theoretical and experimental results, we discuss briefly here what was already done earlier in Ref. [13]. In Ref. [13], the SD method, which is equivalent to the LCCSD method and SD method with important partial triples corrections (SDpT method) were employed to calculate the electric dipole (E1) matrix elements. These values are further tweaked by scaling the wave functions of the SD method (SD_{sc} method) and of the SDpT method (SDpT_{sc} method). Using the E1 matrix elements from the SDpT method and separately combining with the values extracted from the measured lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states of Refs. [16] and with few more E1 matrix elements from the SDpT method, scalar dipole polarizabilities of the $6p^2P_{1/2}$ and $6p^2P_{3/2}$ states of Cs were evaluated. Differential polarizabilities of these $6P$ states with respect to the ground state were determined by taking the precisely measured ground state dipole polarizability as $401.0(6) ea_0^3$ [34], for the Bohr radius a_0 . These differential polarizabilities were then compared with the direct measured values [35–37]. This comparison demonstrated that the values obtained using the E1 matrix elements entirely from the SDpT method match better with the experimental results than the values obtained in the combined approach. On this basis it was argued that the measured values of the lifetimes of the $5D$ states and the differential polarizabilities of the $6P$ states were inconsistent. For this reason it was assumed that the E1 matrix elements obtained using the SDpT method are more accurate. Using the E1 matrix elements from the SD method of Ref. [13], the lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states were ob-

tained as 1114 ns and 1547 ns respectively. These values were changed to 966 ns and 1350 ns for the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states, respectively, when the matrix elements were improved using the SD_{sc} method. Similarly, the lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states were obtained as 1010 ns and 1409 ns respectively upon the use of the matrix elements from the SDpT method. Finally, they were obtained as 981 ns and 1369 ns respectively using the matrix elements from the SDpT_{sc} method.

In the above analysis, two things need to be carefully scrutinized further. First, it can be noticed that after scaling the wave functions the results are changed significantly. This is very difficult to justify from the first principle accuracies in the results by scaling the wave functions. Large differences between the E1 matrix elements obtained before and after scaling the wave functions indicate that it is necessary to consider more physical effects in the above employed SD or SDpT method to improve accuracies in the results. Secondly, it can be found from the sum-over-states approach employed in Ref. [13] to evaluate polarizabilities of the $6P$ states that the principal contributions come from the E1 matrix elements between the $6p^2P_{1/2} \rightarrow 5d^2D_{3/2}$, $6p^2P_{3/2} \rightarrow 5d^2D_{3/2}$ and $6p^2P_{3/2} \rightarrow 5d^2D_{5/2}$ transitions. However, there are also ample amount of contributions come from the E1 matrix elements involving other D excited states. Again, sum-over-states approach has limitations that it cannot account contributions from the core orbitals, higher excited states and continuum accurately. In order to verify accuracies of the theoretically estimated lifetimes of the $5D$ states, we would like to carry out calculations of the E1 matrix elements of the $6p^2P_{1/2} \rightarrow 5d^2D_{3/2}$, $6p^2P_{3/2} \rightarrow 5d^2D_{3/2}$ and $6p^2P_{3/2} \rightarrow 5d^2D_{5/2}$ transitions considering more physical effects than the LCCSD method; especially through the non-linear terms of the CCSD method. However, we also employ other lower order many-body methods such as the Dirac-Hartree-Fock (DHF) method, lower order perturbation theory (MBPT method) and LCCSD method to demonstrate gradual changes in the results with the propagation of the correlation effects through the higher order terms. We also give contributions from the important triples excitations in a variety of procedures and from a semi-empirical approach by using the experimental energies in the calculations of the wave functions in the RCC method. Again, the Dirac-Coulomb (DC) Hamiltonian was considered in Ref. [13]. We also estimate corrections due to the higher order relativistic effects by considering the Breit interaction and lower order quantum electrodynamics (QED) effects in the calculations.

Many general implications for studying the lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states in Cs are already discussed before. We, however, would also like to emphasize on two applications here for which the present work could be directly relevant. The most precise PNC measurement has been carried out in the $6s^2S_{1/2} \rightarrow 7s^2S_{1/2}$ transition of Cs, but the nuclear parameters inferred from the NAM deduced from this measurement are in disagreement with

the values given by the well established nuclear models [7, 8]. This urges for further investigation of PNC effects in the atomic systems. In fact, theoretical study demonstrates the PNC amplitudes in the $6s\ ^2S_{1/2} \rightarrow 5d\ ^2D_{3/2,5/2}$ transitions are almost three times larger than the $6s\ ^2S_{1/2} \rightarrow 7s\ ^2S_{1/2}$ transition in Cs [14, 15]. Plausible principle of measuring the PNC-induced frequency shift in the $6s\ ^2S_{1/2} \rightarrow 5d\ ^2D_{3/2}$ transition of Ba^+ has been described in [38]. Following this, it has also been suggested that the same principle can be adopted to measure PNC effect in the $6s\ ^2S_{1/2} \rightarrow 5d\ ^2D_{3/2}$ transition of Cs [39]. In another work, it has been highlighted that measurement induced light-shifts in the the $S-D_{5/2}$ transitions of the atomic systems would provide unambiguous signature of existence of NAM [40, 41]. One of the requirements to enhance the signal-to-noise ratio in the PNC-induced light-shift measurement principle is to have longer lifetimes of the states involved in the transition [38]. Thus, it is indispensable to ensure reliability in the observed lifetimes of the $5d\ ^2D_{3/2}$ and $5d\ ^2D_{5/2}$ states in case the $6s\ ^2S_{1/2} \rightarrow 5d\ ^2D_{3/2,5/2}$ transitions in Cs are undertaken for the PNC measurement. On the otherhand, it was also advocated that the $5d\ ^2D_{3/2}$ and $5d\ ^2D_{5/2}$ states of Cs are very much suitable for the resonance ionization spectroscopy (RIS) process owing to their longer lifetimes [42]. Therefore, it is important that ambiguity in the correctness of the lifetime values of the $5d\ ^2D_{3/2}$ and $5d\ ^2D_{5/2}$ states of Cs are settled down.

II. THEORY

In the Cs atom, it is obvious to assume that the dominant emission transition probabilities for an electron to jump from the $5d\ ^2D_{3/2}$ and $5d\ ^2D_{5/2}$ states are due to the E1 channel to the low-lying $6p\ ^2P_{1/2}$ and $6p\ ^2P_{3/2}$ states. Since the aim of the present work is to explain the cause of disagreement between the previous theoretical calculations with the experimental results, we intend to show how much the transition probabilities are really small due to the forbidden channels from the above two states. Thus, we also determine transition probabilities due to the next dominant magnetic dipole (M1) and electric quadrupole (E2) channels from the $5d\ ^2D_{3/2}$ state to the ground state $6s\ ^2S_{1/2}$ and from the $5d\ ^2D_{5/2}$ state to the ground and $5d\ ^2D_{3/2}$ states. The general expressions for evaluating these transition probabilities between the $|\Psi_i\rangle \rightarrow |\Psi_f\rangle$ transition are given by

$$A_{if}^{E1} = \frac{2.0261 \times 10^{-6}}{\lambda_{if}^3 g_i} S_{if}^{E1} \quad (1)$$

$$A_{if}^{M1} = \frac{2.6971 \times 10^{-11}}{\lambda_{if}^3 g_i} S_{if}^{M1} \quad (2)$$

and

$$A_{if}^{E2} = \frac{1.1195 \times 10^{-22}}{\lambda_{if}^5 g_i} S_{if}^{E2}, \quad (3)$$

where the quantity $S_{if}^O = |\langle \Psi_i || O || \Psi_f \rangle|^2$ is known as the line strength for the corresponding reduced matrix element $|\langle \Psi_i || O || \Psi_f \rangle|$ of a transition operator O . These quantities are given later in this paper in atomic unit (a.u.). In the above expressions, $g_i = 2J_i + 1$ is the degeneracy factor of the state $|\Psi_i\rangle$ with the angular momentum of the state J_i and the transition wavelength (λ_{if}) is used in nm which when substituted the transition probabilities (A_{if}^O s) are obtained in s^{-1} . The lifetime (τ) of the atomic state $|\Psi_i\rangle$ is determined (in s) by taking reciprocal of the total emission transition probabilities due to all possible channels. i.e.

$$\tau_i = \frac{1}{\sum_{O,f} A_{if}^O}, \quad (4)$$

where the summations over O and f correspond to all the decay channels and all the lower states respectively.

The reduced matrix elements for the E1, M1 and E2 transition operators in terms of the single particle orbitals are given by

$$\begin{aligned} \langle \kappa_f || e1 || \kappa_i \rangle &= \frac{3}{k} \langle \kappa_f || C^{(1)} || \kappa_i \rangle \int_0^\infty dr (j_1(kr) \\ &\times [P_f(r)P_i(r) + Q_f(r)Q_i(r)] + j_2(kr) \\ &\times \left\{ \frac{\kappa_f - \kappa_i}{2} [P_f(r)Q_i(r) + Q_f(r)P_i(r)] \right. \\ &\left. + [P_f(r)Q_i(r) - Q_f(r)P_i(r)] \right\}), \quad (5) \end{aligned}$$

$$\begin{aligned} \langle \kappa_f || m1 || \kappa_i \rangle &= \frac{6}{\alpha k} \frac{(\kappa_f + \kappa_i)}{2} \langle -\kappa_f || C^{(1)} || \kappa_i \rangle \int_0^\infty dr \\ &\times j_1(kr) (P_f(r)Q_i(r) + Q_f(r)P_i(r)) \quad (6) \end{aligned}$$

and

$$\begin{aligned} \langle \kappa_f || e2 || \kappa_i \rangle &= \frac{15}{k^2} \langle \kappa_f || C^{(2)} || \kappa_i \rangle \int_0^\infty dr (j_2(kr) \\ &\times [P_f(r)P_i(r) + Q_f(r)Q_i(r)] + j_3(kr) \\ &\times \left\{ \frac{\kappa_f - \kappa_i}{3} [P_f(r)Q_i(r) + Q_f(r)P_i(r)] \right. \\ &\left. + [P_f(r)Q_i(r) - Q_f(r)P_i(r)] \right\}), \quad (7) \end{aligned}$$

where $P(r)$ and $Q(r)$ denote for the large and small components of the radial parts of the single particle Dirac orbitals, respectively, κ s are their relativistic angular momentum quantum numbers, α is the fine structure constant, $k = \alpha(\epsilon_f - \epsilon_i)$ with the orbital energies ϵ s and $j_l(kr)$ is the spherical Bessel function. The reduced Racah coefficients with rank k are given by

$$\begin{aligned} \langle \kappa_f || C^{(k)} || \kappa_i \rangle &= (-1)^{j_f+1/2} \sqrt{(2j_f+1)(2j_i+1)} \\ &\left(\begin{matrix} j_f & k & j_i \\ 1/2 & 0 & -1/2 \end{matrix} \right) \pi(l_{\kappa_f}, k, l_{\kappa_i}), \quad (8) \end{aligned}$$

with

$$\pi(l, k, l') = \begin{cases} 1 & \text{for } l + k + l' = \text{even} \\ 0 & \text{otherwise,} \end{cases} \quad (9)$$

TABLE I: List of different parameters used to define the basis functions using QTOs in the present calculations.

	s	p	d	f	g
N_l	34	33	32	31	30
η_0	2.0×10^{-8}	2.5×10^{-8}	2.5×10^{-8}	2.1×10^{-1}	2.1×10^{-7}
ζ	4.67	4.78	4.93	7.08	8.25

for the orbital momentum l_κ of the corresponding orbital having the relativistic quantum number κ .

For the first time, we use a different type of analytical basis function having quadratic type of exponents to express the single particle wave functions (define as quadratic type orbitals (QTOs)) to calculate the above reduced matrix elements. Using these functions, the radial components of the orbitals are expressed as

$$P(r) = \sum_{\nu=1}^{N_l} c_\nu^P \mathcal{N}_\nu^P r^l e^{-\eta_\nu r^4}$$

$$\text{and } Q(r) = \sum_{\nu=1}^{N_l} c_\nu^Q \mathcal{N}_\nu^Q r^l \left(\frac{d}{dr} + \frac{\kappa}{r} \right) [r^l e^{-\eta_\nu r^4}], \quad (10)$$

where N_l represents for the total number of QTOs considered in the calculations, η_ν is an arbitrary coefficient suitably chosen to obtain wave functions accurately, $c_\nu^{P(Q)}$ s are the linear combination coefficients, $\mathcal{N}_\nu^{P(Q)}$ is the normalization constant of the ν th basis function for the large (small) component of the wave function. It can be noticed above that the kinetic balance condition between the large and small components has been maintained. The normalization constants are given by

$$\mathcal{N}_\nu^P = 2(2\eta_\nu)^{\frac{2l+1}{8}} \Gamma\left(\frac{2l+1}{4}\right)^{-1/2} \quad (11)$$

and

$$\mathcal{N}_\nu^Q = \left[\frac{(l+\kappa)^2}{4(2\eta_\nu)^{\frac{2l-1}{4}}} \Gamma\left(\frac{2l-1}{4}\right) - \frac{2(l+\kappa)}{(2\eta_\nu)^{\frac{2l-1}{4}}} \right. \\ \left. \times \Gamma\left(\frac{2l+3}{4}\right) + \frac{4}{(2\eta_\nu)^{\frac{2l-1}{4}}} \Gamma\left(\frac{2l+7}{4}\right) \right]^{-1}. \quad (12)$$

For convenience, the η_ν parameters are constructed satisfying the even tempering condition between two parameters η_0 and ζ as

$$\eta_\nu = \eta_0 \zeta^{\nu-1}. \quad (13)$$

We give the list of η_0 and ζ parameters in Table I that are used in the present calculations.

III. MANY-BODY METHODS

In our previous work [20], we have described general procedures of our MBPT(2) and RCC methods using

which we have calculated the wave functions and transition matrix elements of the Fr atom in the approach of Bloch's formalism [23]. We also adopt these methods here along with few more variants of the RCC methods by approximating the levels of excitations and non-linear terms in the expression of the wave function. We apply these methods in order to investigate the reason for the discrepancies between the previous theoretical study with the experimental results [13]. We discuss briefly about these methods below to illustrate distinctly the roles of higher order correlation effects to enhance accuracies in the calculations of the transition matrix elements.

In the Bloch's prescription the atomic wave function of a state $|\Psi_v\rangle$ of Cs with a valence orbital v is expressed as [23]

$$|\Psi_v\rangle = \Omega_v |\Phi_v\rangle, \quad (14)$$

where Ω_v and $|\Phi_v\rangle$ are referred to as the wave operator and the reference state respectively. For the computational simplicity we choose the working reference state as the DHF wave function $|\Phi_c\rangle$ for the closed-shell configuration $[5p^6]$, which is common to the ground and the excited states that are involved in the estimations of the lifetimes of the $5D$ states of Cs. Then, the actual reference state is constructed from it as $|\Phi_v\rangle = a_v^\dagger |\Phi_c\rangle$ for the respective state with the valence orbital v . First the calculations are performed using the DC Hamiltonian which in a.u. is given by

$$H = \sum_i \left[c\alpha_i \cdot \mathbf{p}_i + (\beta_i - 1)c^2 + V_n(r_i) + \sum_{j>i} \frac{1}{r_{ij}} \right], \quad (15)$$

with α and β are the usual Dirac matrices and $V_n(r)$ represents for the nuclear potential. We evaluate the nuclear potential considering the Fermi-charge distribution defined by

$$\rho_n(r) = \frac{\rho_0}{1 + e^{(r-b)/a}}, \quad (16)$$

for the normalization factor ρ_0 , the half-charge radius b and $a = 2.3/4(\ln 3)$ is related to the skin thickness. We have used $a = 2.3/4(\ln 3)$ and $b = 5.6707$ fm, which is determined using the relation

$$b = \sqrt{\frac{5}{3} r_{rms}^2 - \frac{7}{3} a^2 \pi^2} \quad (17)$$

with the root mean square (rms) charge radius of the nucleus determined using the formula

$$r_{rms} = 0.836 A^{1/3} + 0.570 \quad (18)$$

in fm for the atomic mass A .

Contributions from the frequency independent Breit interaction are estimated by adding the corresponding interaction term given by

$$V_B(r_{ij}) = -\frac{1}{2r_{ij}} \{ \alpha_i \cdot \alpha_j + (\alpha_i \cdot \hat{\mathbf{r}}_{ij})(\alpha_j \cdot \hat{\mathbf{r}}_{ij}) \}. \quad (19)$$

We have also estimated lower order quantum electrodynamic (QED) effects by considering the following potentials with H in a similar formalism as described in Ref. [43] but for the above nuclear Fermi-charge distribution. The lower order vacuum polarization (VP) effects are considered at the approximations of Uehling ($V_U(r)$) and Wichmann-Kroll ($V_{WK}(r)$) potentials given by

$$V_U(r) = -\frac{2\alpha^2}{3r} \int_0^\infty dx x \rho_n(x) \int_1^\infty dt \sqrt{t^2 - 1} \\ \times \left(\frac{1}{t^3} + \frac{1}{2t^5} \right) \left[e^{-2ct|r-x|} - e^{-2ct(r+x)} \right] \quad (20)$$

and

$$V_{WK}(r) = -\frac{8Z^2\alpha^4}{9r} (0.092) \int_0^\infty dx x \rho_n(x) \\ \times (0.22 \{ \arctan[1.15(-0.87 + 2c|r-x|)] \\ - \arctan[1.15(-0.87 + 2c(r+x))] \} \\ + 0.22 \{ \arctan[1.15(0.87 + 2c|r-x|)] \\ - \arctan[1.15(0.87 + 2c(r+x))] \} \\ - 0.11 \{ \ln[0.38 - 0.87c|r-x| + c^2(r-x)^2] \\ - \ln[0.38 - 0.87c(r+x) + c^2(r+x)^2] \} \\ + 0.11 \{ \ln[0.38 + 0.87c|r-x| + c^2(r-x)^2] \\ - \ln[0.38 + 0.87c(r+x) + c^2(r+x)^2] \}), \quad (21)$$

with the atomic number of the system Z . The contribution from the self-energy (SE) interaction are accounted by evaluating contributions together from the electric form-factor given by

$$V_{SE}^{ef}(r) = -A(Z)(Z\alpha)^4 e^{-Zr} + \frac{B(Z,r)\alpha^2}{r} \int_0^\infty dx x \rho_n(x) \\ \times \int_1^\infty dt \frac{1}{\sqrt{t^2 - 1}} \left\{ \left(\frac{1}{t} - \frac{1}{2t^3} \right) \right. \\ \times \left[\ln(t^2 - 1) + 4 \ln \left(\frac{1}{Z\alpha} + \frac{1}{2} \right) \right] - \frac{3}{2} + \frac{1}{t^2} \} \\ \times \left[e^{-2ct|r-x|} - e^{-2ct(r+x)} \right] \quad (22)$$

and from the magnetic form-factor given by

$$V_{SE}^{mg}(r) = \frac{i\alpha}{4\pi c} \boldsymbol{\gamma} \cdot \boldsymbol{\nabla}_r \int_0^\infty d^3x \rho_n(x) \\ \times \left[\left(\int_1^\infty dt \frac{e^{-2ctR}}{Rt^2 \sqrt{t^2 - 1}} \right) - \frac{1}{R} \right], \quad (23)$$

where $A(Z) = 0.074 + 0.35Z\alpha$, $B(Z,r) = [1.071 - 1.97((Z - 80)\alpha)^2 - 2.128((Z - 80)\alpha)^3 + 0.169((Z - 80)\alpha)^4]cr/(cr + 0.07(Z\alpha)^2)$ and $R = |\mathbf{r} - \mathbf{x}|$.

Following the form of the reference states in our approach, Ω_v can now be divided as

$$\Omega_v = 1 + \chi_c + \chi_v, \quad (24)$$

where χ_c and χ_v are responsible for carrying out excitations from $|\Phi_c\rangle$ and $|\Phi_v\rangle$, respectively, due to the residual

interaction $V_r = H - H_0$ for the DHF Hamiltonian H_0 . In a perturbative series expansion, we can express as

$$\chi_c = \sum_k \chi_c^{(k)} \quad \text{and} \quad \chi_v = \sum_k \chi_v^{(k)}, \quad (25)$$

where the superscript k refer to the number of times V_r is considered in the MBPT method (denoted by MBPT(k) method). The k th order amplitudes for the χ_c and χ_v operators are obtained by solving the equations [23]

$$[\chi_c^{(k)}, H_0]P = QV_r(1 + \chi_c^{(k-1)})P \quad (26)$$

and

$$[\chi_v^{(k)}, H_0]P = QV_r(1 + \chi_c^{(k-1)} + \chi_v^{(k-1)})P - \sum_{m=1}^{k-1} \chi_v^{(k-m)} \\ \times PV_r(1 + \chi_c^{(m-1)} + \chi_v^{(m-1)})P \quad (27)$$

with $\chi_c^{(0)} = 0$ and $\chi_v^{(0)} = 0$, where the projection operators $P = |\Phi_c\rangle\langle\Phi_c|$ and $Q = 1 - P$ describe the model space and the orthogonal space of the DHF Hamiltonian H_0 respectively. The energy of the state $|\Psi_n\rangle$ is evaluated by using an effective Hamiltonian

$$H_v^{eff} = Pa_v H \Omega_v a_v^\dagger P. \quad (28)$$

Using normal order Hamiltonian $H_N = H - PHP$ in place of H in the above expression, attachment energy of a state with the valence orbital v is evaluated.

In the (R)CC theory ansatz, wave functions of the considered states are expressed as

$$|\Psi_v\rangle \equiv \Omega_v |\Phi_v\rangle = e^T \{1 + S_v\} |\Phi_v\rangle \quad (29)$$

with $\chi_c = e^T - 1$ and $\chi_v = e^T S_v - 1$, where T and S_v are the CC excitation operators that excite electrons from the core and core along with the valence orbitals to the virtual space respectively. In this work, we have considered only the single and double excitations, denoted by the subscripts 1 and 2 respectively, in the CCSD method as

$$T = T_1 + T_2 \quad \text{and} \quad S_v = S_{1v} + S_{2v}. \quad (30)$$

In the LCCSD method only the linear terms are retained as in the SD method of Ref. [13]. The amplitudes of these operators are evaluated using the equations

$$\langle \Phi_c^* | \bar{H}_N | \Phi_c \rangle = 0 \quad (31)$$

and

$$\langle \Phi_v^* | (\bar{H}_N - \Delta E_v) S_v | \Phi_v \rangle = -\langle \Phi_v^* | \bar{H}_N | \Phi_v \rangle, \quad (32)$$

where $|\Phi_c^*\rangle$ and $|\Phi_v^*\rangle$ are the excited state configurations, here up to doubles, with respect to the DHF states $|\Phi_c\rangle$ and $|\Phi_v\rangle$ respectively and $\bar{H}_N = (H_N e^T)_l$ with subscript l represents for the linked terms only. Here

$\Delta E_v = H_v^{eff} - H_c^{eff}$ is the attachment energy of the electron in the valence orbital v with $H_c^{eff} = PH(1 + \chi_c)P$. Following Eq. (28), expression for ΔE_v is given by

$$\Delta E_v = \langle \Phi_v | \bar{H}_N \{1 + S_v\} | \Phi_v \rangle. \quad (33)$$

We also include contributions from the important triply excited configurations by defining perturbative operators defined as

$$T_3^{pert} = \frac{1}{6} \sum_{abc,pqr} \frac{(H_N T_2)_{abc}^{pqr}}{\epsilon_a + \epsilon_b + \epsilon_c - \epsilon_p - \epsilon_q - \epsilon_r}, \quad (34)$$

and

$$S_{3v}^{pert} = \frac{1}{4} \sum_{ab,pqr} \frac{(H_N T_2 + H_N S_{2v})_{abv}^{pqr}}{\epsilon_a + \epsilon_b + \epsilon_v - \epsilon_p - \epsilon_q - \epsilon_r}, \quad (35)$$

where $\{a, b, c\}$ and $\{p, q, r\}$ represent for the occupied and virtual orbitals respectively and ϵ s are their corresponding orbital energies. Since the final results reported in Ref. [13] are using the SDpT method and scaling the wave functions, we would like to find out roles of the triply excited configurations in the evaluation of the transition matrix elements. However the exact procedure using which triple excitations are accounted in the SDpT method is not clear to us, so we try to estimate these contributions in various possible ways. When the S_{3v}^{pert} operator is considered as a part of the S_v operator to estimate only the energies using Eq. (33) after obtaining the RCC amplitudes, it is referred to as (L)CCSD(T) method. However, when it is involved to estimate both the energies and amplitudes of the S_v operators simultaneously in the iterative procedure through Eqs. (32) and (33), we call it as (L)CCSD[T] method. To explore roles of the core correlations through the triple excitations, we consider T_3^{pert} operator as a part of T operator while solving Eq. (31). This is referred to as (L)CCSDpT^c method and when along with this approach, S_{3v}^{pert} operator is considered in Eqs. (32) and (33), we refer to this as (L)CCSDpT method. But, we consider both the T_3^{pert} and S_{3v}^{pert} operators in Eqs. (31) and (32) only to ameliorate amplitudes of the T_1 and S_{1v} operators for the computational easiness.

After obtaining amplitudes of the MBPT and RCC operators using the equations described earlier, the transition matrix element of an operator O between the states $|\Psi_i\rangle$ and $|\Psi_f\rangle$ is evaluated using the expression

$$\frac{\langle \Psi_f | O | \Psi_i \rangle}{\sqrt{\langle \Psi_f | \Psi_f \rangle \langle \Psi_i | \Psi_i \rangle}} = \frac{\langle \Phi_f | \Omega_f^\dagger O \Omega_i | \Phi_i \rangle}{\sqrt{\langle \Phi_f | \Omega_f^\dagger \Omega_f | \Phi_f \rangle \langle \Phi_i | \Omega_i^\dagger \Omega_i | \Phi_i \rangle}}. \quad (36)$$

This gives rise to a finite number of terms for the MBPT(2) and LCCSD-variant methods, but it involves two non-terminating series in the numerator and denominator, which are $e^{T^\dagger} O e^T$ and $e^{T^\dagger} e^T$ respectively, in the CCSD-variant methods. As described in our previous

TABLE II: Demonstration of trends of the calculated energies (in cm⁻¹) using various relativistic methods considered in the present work with the DC Hamiltonian. Relativistic corrections are given separately from the CCSD method. These results are compared with the experimental values [46]. Uncertainties in the experimental values are not mentioned as they are more precise than the quoted values up to the second decimal places. Bold fonts are to highlight accuracies in the results.

Method	6s ² S _{1/2}	6p ² P _{1/2}	6p ² P _{3/2}	5d ² D _{3/2}	5d ² D _{5/2}
DHF	27983.73	18752.17	18350.36	14096.82	14121.80
MBPT(2)	32020.63	20362.23	19777.17	16681.89	16568.09
LCCSD	32425.64	20566.54	19965.95	17882.53	17718.94
LCCSD(T)	31812.61	20335.35	19762.15	17439.99	17325.21
LCCSDpT ^c	32425.66	20566.54	19965.95	17882.53	17718.94
LCCSD[T]	31834.43	20340.48	19766.15	17505.35	17381.71
LCCSDpT	31758.84	20310.25	19741.51	17374.41	17281.31
CCSD	31463.22	20159.54	19600.28	16537.86	16445.08
CCSD(T)	31090.05	20011.04	19470.48	16259.53	16149.22
CCSDpT ^c	31428.69	20149.29	19591.82	16504.24	16414.53
CCSD[T]	31064.13	20013.07	19472.02	16272.98	16214.82
CCSDpT	31001.99	19993.26	19455.22	16223.73	16170.23
Relativistic corrections					
Breit	-0.40	-7.50	-1.32	20.17	23.62
VP	3.63	-0.03	-0.09	-0.40	-0.36
SE	-17.92	-1.09	0.95	2.11	2.15
Breit+QED	-14.86	-8.62	-0.47	21.87	25.42
Experiment	31406.47	20228.20	19674.26	16907.21	16809.62

works [20, 44, 45], we adopt iterative procedures to account contributions from these non-truncative series. To comprehend, we also give intermediate results keeping different k number of T and/or T^\dagger operators in these series of the CCSD method for evaluating the matrix elements and refer to the approach as CCSD^(k) method. Finally, our CCSD results correspond to the calculations using the CCSD^(∞) method. We also estimate contributions due to the triply excitations by considering both the T_3^{pert} and S_{3v}^{pert} operators along with their complex conjugates in Eq. (36) of the (L)CCSD method and refer the approach as (L)CCSD_{t3} method.

IV. RESULTS AND DISCUSSION

Before presenting the transition matrix elements from various methods, we would like to first validate the methods by carrying out calculations of the attachment energies of the considered states of Cs and comparing them against their corresponding experimental values. Although it is understood that accuracies in the radial parts of the wave functions could be different in the accurate evaluation of the energies and transition matrix elements, but it can be noticed from Eqs. (27) and (32) that the

TABLE III: Comparison of E1 reduced matrix elements (in a.u.) from various methods. Relativistic corrections are quoted separately and our recommended values are given as “Reco”. Results from other recent calculations are also given.

Method	$5d_{3/2} \rightarrow 6p_{1/2}$	$5d_{3/2} \rightarrow 6p_{3/2}$	$5d_{5/2} \rightarrow 6p_{3/2}$
DHF	9.012	4.078	12.233
MBPT(2)	7.535	3.404	10.273
LCCSD	6.566	2.954	9.011
LCCSD _{t3}	6.569	2.952	9.015
LCCSD _{pT} ^c	6.566	2.954	9.011
LCCSD[T]	6.472	2.909	8.899
LCCSD _{pT}	6.687	3.009	9.137
LCCSD _{ex}	6.305	2.828	8.683
CCSD ⁽²⁾	7.292	3.291	9.931
CCSD ⁽⁴⁾	7.301	3.295	9.941
CCSD^(∞)	7.301	3.295	9.941
CCSD _{t3}	7.304	3.293	9.945
CCSD _{pT} ^c	7.326	3.307	10.018
CCSD[T]	7.258	3.275	9.934
CCSD _{pT}	7.357	3.320	10.056
CCSD _{ex}	7.348	3.318	10.050
Relativistic corrections			
Breit	−0.009	−0.005	0.022
VP	~ 0.0	~ 0.0	0.039
SE	−0.001	−0.001	0.037
Breit+QED	−0.010	−0.005	0.020
Estimated Uncertainties			
Basis	0.048	0.023	0.022
Triples	0.003	0.002	0.004
Scaling	0.047	0.023	0.109
Recommended values			
Reco	7.291(67)	3.288(33)	9.961(111)
From Ref. [13]			
DHF	8.9784	4.0625	12.1865
MBPT(3)	6.9231	3.1191	9.4545
SD	6.5809	2.9575	9.0238
SD _{sc}	7.0634	3.1871	9.6588
SD _{pT}	6.9103	3.1112	9.4541
SD _{pT} _{sc}	7.0127	3.1614	9.5906
From Ref. [14]			
Σ ⁽²⁾	6.744	3.037	9.254
λΣ ⁽²⁾	7.039	3.173	9.629
Σ ^(∞)	6.927	3.121	9.481
λΣ ^(∞)	7.032	3.170	9.616

energy evaluating expressions are also coupled with the wave function determining equations. Hence, accurate evaluation of the energies using a method can be an indication of the validation of the method in addition to embodying more physical effects in the method. For this purpose, we give energies obtained from various meth-

ods, that are described before, in Table II using the DC Hamiltonian and compare them with the experimental values [46]. We find the CCSD method gives rise fairly accurate results for all the states being considered. It is also noticed that the MBPT(2) values are more accurate than the LCCSD values, but the partial triples effects bring down the LCCSD results closer to the experimental values. However when these partial effects contributions are added in the CCSD results, the results become far off from the experimental results. It, therefore, implies that the neglected triples effects, mainly that can contribute through the T_2 and S_{2v} amplitude determining equations, may cancel out some of these over estimated triple excited contributions to give finally more precise results. We also observe that the triples effects through the valence orbital excitations are the dominant ones over the core-triple excitations. Nevertheless, it would be pertinent to consider full triple excitations in this situation than adapting through the partial effects. Therefore, we consider results from the CCSD method, that accounts all the non-linear terms within the considered level of excitations, as the recommended calculated values for the further use.

We have also explicitly estimated the contributions due to the Breit interaction (given as “Breit”), the VP effect (given as “VP”) and the SE effect (given as “SE”) using the CCSD method which are given towards the bottom of Table II. In addition, we also determine these corrections considering all these relativistic corrections together with respect to the contributions from the DC Hamiltonian in the CCSD method (given as “Breit+QED”). We find slight changes in the results between the “Breit+QED” approach and when the corrections estimated independently are added-up. We also observe that among all these relativistic corrections, the SE effect is large in the ground state while the Breit interaction gives larger corrections in the other considered states.

After analyzing energies from various methods with respect to the experimental values, we also give the E1 matrix elements in Table III of the transitions that are required to estimate the lifetimes of the $5d\ ^2D_{3/2}$ and $5d\ ^2D_{5/2}$ states of Cs. We compare these results with the values reported recently by the other groups using different relativistic many-body methods [13, 14]. In addition to the methods we have employed to evaluate the energies, we also give E1 matrix elements using the (L)CCSD_{t3} and (L)CCSD_{ex} methods in the above table. We find reasonable agreements between the results obtained using our DHF and LCCSD methods with the DHF and SD methods of Ref. [13]. However, there are significant differences in the results when the higher order effects are accounted. Similarly, our results differ substantially from the calculations reported in Ref. [14] in which a combination of correlation potential (CP) method (k th order is denoted by $\Sigma^{(k)}$) and time-dependent Hartree-Fock (TDHF) method with the Brueckner orbitals (BOs) are employed. Moreover, the E1 matrix elements quoted in Ref. [13] are improved

TABLE IV: Reduced matrix elements (in a.u.) due to the E2 and M1 transitions given from different methods. Relativistic corrections and Reco values along with the uncertainties are given at the end of the table. The most accurate calculations are highlighted by the bold fonts.

Method	$5d_{3/2} \rightarrow 6s_{1/2}$		$5d_{5/2} \rightarrow 6s_{1/2}$	$5d_{5/2} \rightarrow 5d_{3/2}$	
	M1	E2	E2	M1	E2
DHF	~ 0.0	43.844	53.707	1.549	44.287
MBPT(2)	3.2×10^{-5}	34.287	42.217	1.549	29.127
LCCSD	8.8×10^{-5}	31.149	38.642	1.547	23.774
LCCSD _{t3}	8.8×10^{-5}	31.165	38.623	1.547	23.782
LCCSD _{pT} ^c	8.8×10^{-5}	31.149	38.642	1.547	23.774
LCCSD[T]	8.9×10^{-5}	30.698	38.153	1.547	23.033
LCCSD _{pT}	8.7×10^{-5}	31.979	39.541	1.548	24.497
LCCSD _{ex}	9.1×10^{-5}	30.053	37.436	1.548	21.741
CCSD ⁽²⁾	7.6×10^{-5}	35.301	43.437	1.547	28.878
CCSD ⁽⁴⁾	2.1×10^{-4}	35.331	43.468	1.551	28.897
CCSD ^(∞)	2.2×10^{-4}	34.400	42.441	1.551	29.037
CCSD _{t3}	2.2×10^{-4}	34.416	42.422	1.551	29.045
CCSD _{pT} ^c	2.1×10^{-4}	34.523	42.598	1.551	29.248
CCSD[T]	2.2×10^{-4}	34.172	42.203	1.551	28.620
CCSD _{pT}	2.0×10^{-4}	34.897	42.960	1.551	29.323
CCSD _{ex}	2.2×10^{-4}	34.530	42.600	1.551	29.622
Relativistic corrections					
Breit	~ 0.0	-0.031	-0.047	~ 0.0	-0.105
VP	~ 0.0	-0.004	-0.004	~ 0.0	0.003
SE	~ 0.0	0.019	0.024	~ 0.0	-0.012
Breit+QED	~ 0.0	-0.015	-0.028	~ 0.0	-0.116
Estimated Uncertainties					
Basis	~ 0.0	0.106	0.112	~ 0.0	0.289
Triples	2.0×10^{-5}	0.016	0.019	0.001	0.008
Scaling	~ 0.0	0.130	0.159	~ 0.0	0.585
Recommended values					
Reco	$2.2(2) \times 10^{-4}$	34.385(168)	42.413(195)	1.550(1)	28.921(653)

drastically using the SD_{sc} and SD_{pT}_{sc} methods where the wave functions are scaled to account the omitted contributions. Large differences in the results obtained before and after scaling the wave functions demand for including the omitted contributions more accurately. In Ref. [14] too, the final results are quoted using the $\lambda\Sigma^{(k)}$ approach with the scaling parameter λ . Our CCSD method includes more physical effects through its formulation [23–25] and this is also partly justified from the comparison of energies in Table II. Therefore, we consider results from the CCSD method as more reliable since it includes all the non-linear terms within the considered levels of approximations and accounts pair-correlation and core-polarization effects to all orders [22]. These non-linear terms take care of most of the contributions from the triple and quadrupole excitations; more importantly both the singly and doubly excited amplitudes see these

effects equitably. To show the effectiveness of these non-linear terms, we also evaluate the E1 matrix elements considering the same linear form of the RCC terms in Eq. (36) through our CCSD⁽²⁾ method that naturally appears in the LCCSD method. As seen in Table III, the differences between the results from the LCCSD and CCSD⁽²⁾ methods are quite large. This countenances our above assertion. Compared to the results from the MBPT(2) method, results obtained using the (LCC)SD and MBPT(3) methods from our calculations and from Ref. [13] are smaller but the CCSD values are closer. This means there are strong cancellations in the correlation effects among the higher order terms. This trend is similar to the calculations of energies as seen in Table II. We also notice amount of contributions estimated through the partial triples effects by us and given in Ref. [13] are very different. This may be owing to the fact

TABLE V: Transition wavelengths (in nm) and probabilities (A_{if}^O) due to different decay channels (O s) in s^{-1} from the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states of Cs from various works. Uncertainties are quoted within the parentheses. We also compare our results with the available other theoretical and experimental values. Results only from the SDpT_{sc} method are quoted from Ref. [13].

Transition	λ_{if} (nm)	O	A_{if}^O (in s^{-1})		τ_i (in ns)		
$J_i \rightarrow J_f$	[46]		This work	Others	This work	Others	Experiment
$5d^2D_{3/2} \rightarrow 6p^2P_{1/2}$	3011.15	E1	986229(18209)	804000 [13]	907(16)	981 [13]	909(15) [16]
$5d^2D_{3/2} \rightarrow 6p^2P_{3/2}$	3613.96	E1	116015(2341)	94000 [13]		909 [29]	890(90) [26]
$5d^2D_{3/2} \rightarrow 6s^2S_{1/2}$	689.69	E2	21.21(20)			1061 [30]	1250(115) [27]
$5d^2D_{3/2} \rightarrow 6s^2S_{1/2}$		M1	~ 0			952 [31]	
						970 [32]	
						856 [33]	
$5d^2D_{5/2} \rightarrow 6p^2P_{3/2}$	3490.84	E1	787636(17652)	646000	1270(28)	1369 [13]	1281(9) [16]
$5d^2D_{5/2} \rightarrow 6s^2S_{1/2}$	685.08	E2	22.24(21)			1283 [29]	1225(12) [17]
$5d^2D_{5/2} \rightarrow 5d^2d_{3/2}$	102469.52	E2	~ 0			1434 [30]	890(90) [26]
$5d^2D_{5/2} \rightarrow 5d^2d_{3/2}$		M1	~ 0			1370 [31]	1250(115) [27]
						1342 [32]	1260(80) [28]
						1190 [33]	

that triples effects are incorporated differently in both the works. Differences between the LCCSD and LCCSDpT results in our calculations are larger than the difference between the CCSD and CCSDpT results. It means partial triples effects change results in the LCCSD approximation more than the CCSD method. In contrast, we find the differences between the LCCSD and LCCSDpT^c results are much smaller than the differences between the CCSD and CCSDpT^c results implying core-correlations enhance through the non-linear terms of the RCC theory. We also observe relatively small changes in the results obtained using the CCSD⁽²⁾, CCSD⁽⁴⁾ and CCSD^(∞) approximations. Thus, the roles of the non-linear terms of the CCSD method are more effective in the determination of the wave functions than the property evaluation. We also observe both the Breit and QED corrections are of decent size for determining precise value of the E1 matrix element of the $5d^2D_{5/2} \rightarrow 6p^2P_{3/2}$ transition. In fact it is interesting to note here that, unlike in the energy calculations, total sum of the relativistic corrections to the above E1 matrix element obtained from the Breit interaction, VP effect and SE effect are quite different than when they are estimated considering all the interactions (Breit+QED approach) together in the CCSD method. In the other transitions, these corrections are found to be mere in magnitude.

In order to satisfactorily address issues related to the inconsistencies between the previously estimated theoretical results for the lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states of Cs with the experimental values, it is also essential to estimate uncertainties associated with the E1 matrix elements carefully. Obviously, it can be argued that our calculations have uncertainties from three major sources: (a) use of finite basis size, (b) approximations in the levels of excitations in the RCC theory and (c) *ab initio* approach for calculating the wave functions.

Among these three, the first two sources of uncertainties are quite understandable. To fathom about the uncertainty due to the *ab initio* approach, one can follow from Eq. (32) that both the wave function and energy determining equations are coupled. Therefore, uncertainties associated in both the solutions either may be canceled out each other or will be added-up in the final evaluation. If the experimental energy is used in Eq. (32) (which may be treated as a semi-empirical approach) then the uncertainty associated with the energy can be removed (assuming that the experimental energy is more precise). We estimate uncertainties due to the truncated basis size (given as “Basis”) by carrying out calculations with the high lying orbitals using the MBPT(2) method that are neglected in the RCC calculations to circumvent the computational limitations. Uncertainty due to the neglected triples (given as “Triples”) are accounted by taking differences between the results obtained using the CCSD and CCSD_{t3} methods. For estimating uncertainties due to the *ab initio* calculations (given as “Scaling”), we consider differences between the results from the CCSD and CCSD_{ex} methods. It is worth mentioning here that most of the partial triples effects seen in our calculations are present inherently within the above estimated differences. Therefore, it takes into account almost all possible major uncertainties of our calculations using the CCSD method. By accounting all these uncertainties in quadrature, we give the recommended values (quoted as “Reco”) of the E1 matrix elements towards the end of Table III. The absolute values are given after adding the relativistic corrections to the CCSD results.

It can be assumed that contributions from the forbidden transition probabilities to the estimations of the lifetimes of the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states of Cs are negligibly small. However, it is necessary to demonstrated in the scenario when there are inconsistencies

among the theoretical and experimental results. For this purpose, we also estimate these quantities explicitly for the lower order M1 and E2 forbidden channels. We give these forbidden transition amplitudes from the $5d\ ^2D_{3/2}$ and $5d\ ^2D_{5/2}$ states in Table IV using the same methods that were employed to calculate the E1 matrix elements. We also give uncertainties to these quantities adopting the same procedure described in the previous paragraph. The trends of these matrix elements from different many-body methods are almost similar to the E1 results except for the M1 matrix element between the $5d\ ^2D_{3/2} \rightarrow 6s\ ^2S_{1/2}$ transition; which is anyway found to be negligibly small. The relativistic corrections are also found to be quite small. The recommended values are given at the end of the table following the same procedure as were given for the E1 matrix elements in Table III.

Using the recommended transition matrix elements given in Tables III and IV and the experimental wavelengths, quoted in Table V from the database [46], we determine the transition probabilities due to all the considered channels from the $5d\ ^2D_{3/2}$ and $5d\ ^2D_{5/2}$ states of Cs. These values are quoted in Table V along with their uncertainties and compared against the values due to the E1 channel obtained using the SDpT_{sc} procedure of Ref. [13]. We find large differences between the results from both the works. From the total polarizabilities of these results, we find the lifetime of the $5d\ ^2D_{3/2}$ state is 907(16) ns against 981 ns reported in Ref. [13]. This is in quite good agreement with the experimental value 909(15) ns reported in Ref. [16]. Similarly, we obtain lifetime of the $5d\ ^2D_{5/2}$ state as 1270(28) ns against 1369 ns of Ref. [13]. Our result again agrees well with the experimental value 1281(9) ns reported in Ref. [16]. In Table V, we also quote estimated lifetimes of these states from some of the previous theoretical and other experimental results. Most of these theoretical estimations were carried out using the non-relativistic theory [29–33]. Nevertheless, theoretically estimated values in Ref. [29] are very close to our values and the experimental results of [16]. Other theoretical results are far away from our calculations. Other experimental values for the lifetimes of the above $5D$ states also have large error bars [26–28]

except for the $5d\ ^2D_{5/2}$ state as 1225(12) ns reported in Ref. [17]. This lies outside the range of the error bar of the value reported in Ref. [16]. Since we have overestimated the uncertainties in our theoretical analysis to provide more reliable results, we anticipate that error bars in our calculations would be smaller than what have been actually reported. From this point of view, our results support experimental values of the lifetimes of the $5D$ states of Cs reported in Ref. [16]. Our calculations also demonstrate that branching ratios of an electron to jump from the $5d\ ^2D_{3/2}$ state to the lower $6p\ ^2P_{1/2}$ is about 90% while to the $6p\ ^2P_{3/2}$ state is about 10%. On the otherhand, an electron can jump from the $5d\ ^2D_{5/2}$ to the $6p\ ^2P_{3/2}$ with almost 100% probability.

V. CONCLUSION

We have employed a variety of relativistic many-body methods mostly in the coupled-cluster theory framework to calculate the energies and transition matrix elements due to both the allowed and forbidden decay channels of the $5d\ ^2D_{3/2}$ and $5d\ ^2D_{5/2}$ states in Cs. Trends in the results from these methods are discussed and importance of considering the non-linear terms for accurate determination of the matrix elements are highlighted. Corrections due to both the Breit interaction and lower order QED effects in these quantities are demonstrated explicitly. Earlier reported inconsistencies between the theoretical and experimental values of the lifetimes of the above states seem to be resolved. Branching ratios due to various channels are also given. Though the forbidden transition probabilities are found to be extremely small, however our calculated values can be quite useful if the proposed measurements of parity non-conservation effects in the $6s\ ^2S_{1/2} \rightarrow 5d\ ^2D_{3/2,5/2}$ transitions in Cs take place in future.

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